

## Arsenate Removal by Coagulation Using Iron Salts and Organic Polymers

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### Abstract

Arsenic occurs naturally in the water in many parts of the world. The coagulation and flocculation method is used for arsenate removal from drinking water using ferric chloride, ferric sulfate, and ferrous sulfate. The effects of coagulant type and dosage on removal efficiency of arsenate are determined by considering the residual iron after sedimentation and filtration in the effluent. At the same time the impact of the type (cationic, anionic, and nonionic) and dosage of organic polymers on the removal efficiency of arsenate was investigated. Residual arsenate concentrations were decreased below 10 mg L<sup>-1</sup> for ferric chloride and ferrous sulfate concentration of 30 and 80 mg L<sup>-1</sup>, respectively. Ferric chloride provided minimum and acceptable residual iron concentration (175 mg L<sup>-1</sup>) after sedimentation and filtration and it was determined as the most effective and economic coagulant type because it requires lower amounts than the others and produces minimum residual iron. The addition of cationic polyelectrolyte aided maximum arsenate removal efficiency although all types of polymers increased the efficiency of the treatment method. The percentage of removal reached a maximum level at the cationic polyelectrolyte concentration of 2.5, 2, and 3 mg L<sup>-1</sup> for ferric chloride, ferric sulfate, and ferrous sulfate, respectively.

**Keywords:** Arsenate removal, coagulation, iron salts, organic polymers, residual iron.

### Demir Tuzları ve Organik Polimer Kullanılarak Koagülasyon Yöntemi ile Arsenat Giderimi Özet

Arsenik dünyanın birçok bölgesinde doğal olarak suda bulunan bir elementtir. Bu çalışmada içme sularından arsenat giderimi için koagülant olarak demir klorür, demir(II) sülfat ve demir(III) sülfatın denendiği koagülasyon ve flokülasyon yöntemi kullanılmıştır. Arıtmadan sonra suda kalan demir konsantrasyonu da göz önünde bulundurularak, arsenat giderme verimi üzerine koagülant türü ve dozunun etkisi araştırılmıştır. Aynı zamanda kullanılan organik polimerlerin türünün (katyonik, anyonik ve noniyonik) ve dozunun da arsenat giderme verimi üzerine etkisi belirlenmiştir. Arıtmadan sonra suda kalan arsenat konsantrasyonu demir klorür için 30 mg L<sup>-1</sup>, demir(II) sülfat için ise 80 mg L<sup>-1</sup> konsantrasyonlarda 10 µg L<sup>-1</sup>'nin altına düşürülebilmektedir. Daha az miktarda kullanılması ve arıtmadan sonra suda kalan minimum demir konsantrasyonunu (175 mg L<sup>-1</sup>) sağlaması açısından demir klorür en etkili ve ekonomik koagülant türü olarak belirlenmiştir. Kullanılan tüm organik polimer türleri arsenat giderme verimini bir miktar arttırmasına rağmen maksimum arsenat giderme verimi katyonik polielektrolit ilavesi ile sağlanmıştır. Demir klorür ile 2.5, demir(III) sülfat ile 2 ve demir(II) sülfat ile 3 mg L<sup>-1</sup> katyonik polielektrolit konsantrasyonlarında arsenat giderme verimi en yüksek seviyeye ulaşmıştır.

**Anahtar Kelimeler:** Arsenat giderimi, demir tuzları, kalıntı demir, koagülasyon, organik polimer.

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### INTRODUCTION

Arsenic is an ubiquitous element found in the atmosphere, soils and rocks, natural waters and organisms. It is mobilised through a combination of natural processes such as weathering reactions,

biological activity, and volcanic emissions as well as through a range of anthropogenic activities (Smedley and Kinniburgh 2002). The presence of elevated levels of arsenic in groundwater has become a major concern all over the world

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especially in Bangladesh and India (Bhattacharyya et al. 2003, Ali 2006, Harvey et al. 2006). Unfortunately, elevated concentrations of arsenic are found in the groundwater which is used as a drinking water source in Turkey. Especially in Western Turkey where high arsenic concentrations in groundwaters (20-3000 mg L<sup>-1</sup>) have been found which is related to the borate deposits, and volcanic formations (Çolak et al. 2003, Çöl and Çöl 2004, Gemici and Tarcan 2004). Long-term uptake of arsenic contaminated water can lead to cardiovascular disease, various types of skin lesions, neurological effects, peripheral vascular disease, respiratory disease, and cancer of the skin, bladder, lung, and liver (Rodriguez et al. 2003, Yoshida et al. 2004, Duker et al. 2005). In order to minimize the health risks, the U.S. Environmental Protection Agency (USEPA) and the World Health Organization (WHO) have reduced the maximum contaminant level (MCL) for arsenic in drinking water from 50 µg L<sup>-1</sup> to 10 µg L<sup>-1</sup> (Lee et al. 2003). The MCL has also been lowered to 10 mg L<sup>-1</sup> in Turkey (Anonymous 2005).

The toxicity of arsenic depends on its binding form. Organic arsenic compounds are less toxic than inorganic arsenic compounds (Bissen and Frimmel 2003). Inorganic arsenic exists in water primarily as oxyanions with oxidation states of 3+ (trivalent-arsenite) and 5+ (pentavalent-arsenate) (Lytle et al. 2004), depending on the type and amounts of sorbents, pH, redox potential (Eh), and microbial activity (Wang and Mulligan 2006).

Arsenic removal technologies for drinking water include physico-chemical processes such as coagulation/filtration and lime softening, adsorption processes, ion exchange processes, membrane filtration, such as reverse osmosis, and electro dialysis reversal (Anonymous 2000). Coprecipitation of arsenic with iron and aluminum salts is a practical and effective treatment process used for removing arsenic from drinking waters (Song et al. 2006, Violante et al. 2006). Arsenite removal during coagulation with aluminum and ferric salts has been shown to be less efficient than arsenate (Hering et al. 1997, Gregor 2001, Han et al. 2002), since under reducing conditions at a pH less than about 9.2, the uncharged arsenite species will predominate (Smedley and Kinniburgh 2002). Therefore oxidation prior to coagulation is necessary to convert arsenite to arsenate species for

better removal (Anonymous 2000).

The performance of coagulation and flocculation is dependent on several factors such as source water characteristics, pH, coagulant dose and type, mixing conditions, and time. Coagulation aids are sometimes used to achieve optimum conditions for coagulation and flocculation. The aim is to obtain faster floc formation, produce denser and stronger flocs, decrease the coagulant dosage, broaden the effective pH band, and improve the removal of turbidity and other impurities (Wang et al. 2005). There are numerous papers about arsenic removal from drinking water by coagulation where only a limited number of studies have examined the use of cationic (Zouboulis and Katsoyiannis 2002, Han et al. 2002, Wickramasinghe et al. 2004) and anionic (Zouboulis and Katsoyiannis 2002) polymers for increasing the removal of arsenic. No studies on arsenic removal by non-ionic polyelectrolyte have been found. On the other hand residual iron after sedimentation and filtration is the most important parameter which indicates the failure of the coagulation process.

The major objective of this study is to investigate the removal efficiencies of arsenate from tap water by coagulation and flocculation with ferric chloride, ferric sulfate, and ferrous sulfate as coagulants and organic polymers as a coagulant aid. The effects and efficiency of using a low amount of cationic, anionic, and nonionic polyelectrolytes on the removal of arsenic was compared. The residual concentration of iron after sedimentation and filtration was monitored for the determination of efficiency of the coagulation process.

## MATERIAL AND METHODS

### Materials

An arsenate solution was prepared from sodium arsenate (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) (purchased from Sigma) and dissolved in tap water to obtain a concentration of 2 mg L<sup>-1</sup>. In the experimental studies, this stock arsenate solution was used after being diluted. The characterization of the tap water used in the experiments is listed in Table 1. Ferric chloride (FeCl<sub>3</sub>·6H<sub>2</sub>O), ferric sulfate (Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·5H<sub>2</sub>O), and ferrous sulfate (FeSO<sub>4</sub>·7H<sub>2</sub>O) were used as coagulants and purchased from Merck and Carlo Erba. The stock solutions of the coagulants (5000 - 10000 mg L<sup>-1</sup>) were prepared by dissolving an appropriate amount in deionised water. The polymers used were MAGNAFLOC LT22 (cationic

polyelectrolyte), MAGNAFLOC LT27 (anionic polyelectrolyte), and MAGNAFLOC LT20 (nonionic polyelectrolyte) obtained from Ciba Speciality (Bradford, England). Analytical reagent grade chemicals were used for the preparation of all solutions and all glassware was acid-washed. The pH of the water was adjusted using diluted hydrochloric acid (HCl) and sodium hydroxide (NaOH) solutions.

#### Coagulation Experiments

Coagulation was performed using the jar test method with 3 min of rapid mixing at  $120 \pm 2$  rpm and 30 min of slow mixing at  $45 \pm 2$  rpm, and afterward the samples were allowed to settle for 30 min. A series of jar tests were performed using the one liter of tap water with  $50 \text{ mg L}^{-1}$  of arsenate concentration. Prior to the addition of the coagulant, the pH was adjusted. At the end of the settling period, to detect the residual arsenic, the samples were filtered using  $0.45\text{-}\mu\text{m}$  pore size membrane filter, and stored for analysis with the addition of HCl to obtain a pH value of 2 in order to conserve the samples until arsenic was detected. All the experiments were performed in duplicate to evaluate test reproducibility under identical conditions and all values represent the average of the duplicate experiments.

#### Analytical Methods

The arsenic analysis of the influent and effluent acidified waters and determination of iron concentrations was made using the hydride generation procedure coupled with inductively coupled plasma-atomic emission spectrometry (HG-ICP-AES) (Optima 2100 DV). The sodium borohydride reduction procedure instantaneously converts  $\text{As}^{3+}$  to arsine gas. However, at room temperature, the reduction of  $\text{As}^{5+}$  to arsine occurs relatively slow. Therefore, total arsenic determination requires a prereductant such as KI to convert all arsenic to the  $3+$  oxidation state prior to the arsine formation step (Anonymous 1998, Guerin et al. 2000). Water was first mixed with a 2 ml mixture of KI (5%), ascorbic acid (5%), and 6 ml HCl (10%) for 30 minutes in a dark place to reduce  $\text{As}(5+)$  to  $\text{As}(3+)$ . Then, the 10 ml solution was taken for analysis of the  $\text{As}(3+)$  concentrations.

The use of arsine gas formation provides both a way to separate the analyte from potential chemical interferences in the sample and to preconcentrate to improve analytical sensitivity. In HG techniques,

**Table 1.** The typical water quality characteristics used for the experiments.

Parameter	Concentration
pH	8.2
Turbidity, NTU	0.1
Chloride, $\text{mg Cl L}^{-1}$	46
Nitrate, $\text{mg N L}^{-1}$	3
Iron, $\text{mg L}^{-1}$	0.04
Manganese, $\text{mg L}^{-1}$	0.0141
Sodium, $\text{mg L}^{-1}$	23
Conductivity, $\mu\text{S cm}^{-1}$	463
Sulfate, $\text{mg L}^{-1}$	36

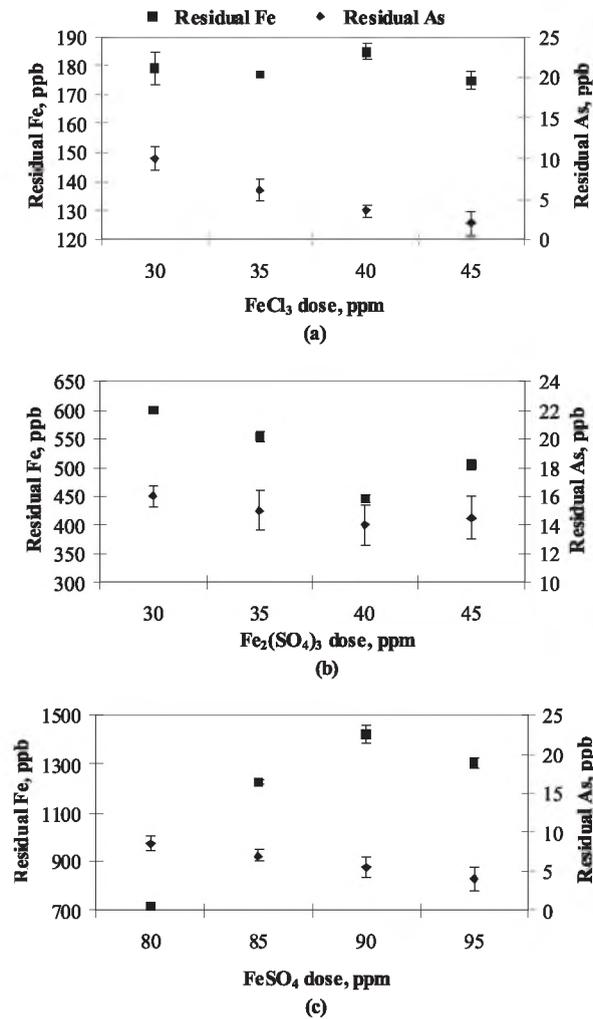
only gaseous hydrides are introduced to the detector and the remaining sample matrix is discarded. As a result, chemical interferences are eliminated (Anonymous 1998, Guerin et al. 2000).

## RESULTS

### Arsenic Removal by Coagulation with Ferric ions

The removal of arsenate from drinking water was investigated by the coagulation and flocculation method using ferric chloride, ferric sulfate, and ferrous sulfate. Firstly, the effects of the coagulant type and dosage on removal efficiency of arsenate was determined by considering the residual iron after sedimentation and filtration in the effluent. The pH of the samples was adjusted as 7.5, 8, and 9 for ferric chloride, ferric sulfate, and ferrous sulfate, respectively. These pH values were selected based on values obtained from relevant studies given in literature (Han et al. 2002, Sancha 2006, Bilici Baskan and Pala 2009). The effect of iron dosages on the arsenic removal efficiency and relationship between coagulant dose and residual iron is shown in Figure 1(a-c).

As it can be seen, after the coagulation and flocculation experiments using ferric chloride and ferrous sulfate, residual arsenate concentrations decreased to below  $10 \mu\text{g L}^{-1}$  which is the maximum contaminant level of arsenic in drinking water determined by the USEPA and WHO. However, this efficiency was obtained for ferric chloride at concentration of  $30 \text{ mg L}^{-1}$ , while it was provided at



**Fig 1.** The effect of a) ferric chloride b) ferric sulfate, and c) ferrous sulfate on the arsenic removal efficiency and relationship between coagulant dose and residual iron.

a coagulant dose of 80 mg L<sup>-1</sup> for ferrous sulfate. Maximum arsenate removal efficiency was achieved as 74% at an Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> concentration of around 40 mg L<sup>-1</sup>.

The effect of ferric chloride, ferric sulfate, and ferrous sulfate dosages on iron residuals is shown in Figure 1(a-c). Minimum residual iron concentration was measured as 175, 446, and 716 μg L<sup>-1</sup> for ferric chloride, ferric sulfate, and ferrous sulfate, respectively. Because the maximum contaminant level of iron in drinking water is 200 μg L<sup>-1</sup> according to the Turkish Standards (Anonymous 2005), ferric chloride proved to be a more effective and reliable coagulant than others used in the present study considering the required

concentration and residual iron.

**Effect of Organic Polymers on Arsenic Removal During Coagulation**

In order to determine the effect of organic polymers on the removal efficiencies of arsenate from tap water by the coagulation and flocculation method using ferric chloride, ferric sulfate, and ferrous sulfate as coagulants, some experiments were performed and the impact of the polyelectrolyte type (cationic, anionic, and nonionic) and dosage was determined. Firstly, the effect of the ferric chloride and polymers on arsenate removal efficiency at the initial arsenate concentration of 50 μg L<sup>-1</sup> and at a pH of 7.5 was investigated. Figure 2 depicts the variations of percent arsenate removal at a FeCl<sub>3</sub> concentration of 25 mg L<sup>-1</sup> by coagulation enhanced with an anionic, cationic, or nonionic polyelectrolyte.

Using FeCl<sub>3</sub> as a coagulant without the addition of a polymer yielded 72% arsenate removal. The percent of arsenate removal generally increased with the increasing concentrations of cationic, anionic, and nonionic polyelectrolyte content up to 1.5 mg L<sup>-1</sup> for cationic and anionic and 2.5 mg L<sup>-1</sup> for nonionic polyelectrolyte and when the cationic and anionic polyelectrolyte dose went above 1.5 mg L<sup>-1</sup> arsenate removal efficiency did not change. The percent of arsenate removal reached a maximum level at a cationic polyelectrolyte concentration of 2.5 mg L<sup>-1</sup>. Increasing the coagulant aid dose from 0.25 to 2.5 mg L<sup>-1</sup> resulted in an increase in arsenate removal efficiency from 74 to 76%. Addition of anionic and nonionic polyelectrolyte provided a maximum of 75 and 74% arsenate removal, respectively.

The variations of the percent of arsenate removals with a cationic, anionic, and nonionic polyelectrolyte dose at a constant pH of 8 and at a ferric sulfate dose of 40 mg L<sup>-1</sup> are shown in Figure 3. Ferric sulfate concentration of 40 mg L<sup>-1</sup> obtained 72% arsenate removal without the addition of a polymer as a result of precipitation, co-precipitation and adsorption mechanisms. Increasing the cationic and anionic polyelectrolyte doses from 0.4 to 2 mg L<sup>-1</sup> and increasing the nonionic polyelectrolyte dose from 0.4 to 4 mg L<sup>-1</sup> caused an increase in the arsenate removal efficiency. Arsenate removal efficiency did not change above 2 mg L<sup>-1</sup> of both the cationic and anionic polyelectrolyte just as is the case of the ferric chloride. The percent of arsenate

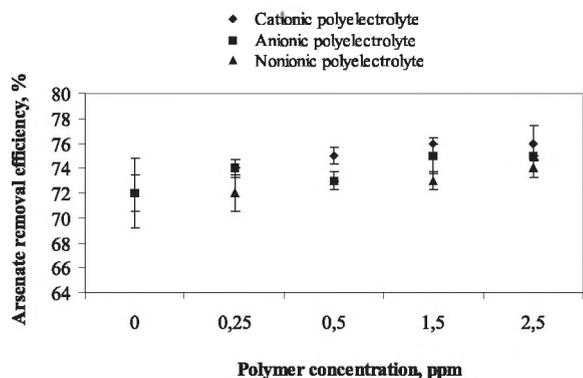


Fig 2. The effect of polyelectrolyte addition on the performance of the coagulation process with  $\text{FeCl}_3$  ( $25 \text{ mg L}^{-1}$ )

removal reached a maximum level at a cationic polyelectrolyte concentration of  $2 \text{ mg L}^{-1}$ . Increasing the coagulant aid dose from  $0.4$  to  $2 \text{ mg L}^{-1}$  resulted in an increase in arsenate removal efficiency from  $75$  to  $76\%$ . Addition of both anionic and nonionic polyelectrolyte provided a maximum of  $75\%$  arsenate removal.

Figure 4 depicts the variations of percent arsenate removal at a constant pH of  $9$  and constant  $\text{FeSO}_4$  concentration of  $60 \text{ mg L}^{-1}$  by coagulation enhanced with anionic, cationic, or nonionic polyelectrolyte. The  $\text{FeSO}_4$  dose of  $60 \text{ mg L}^{-1}$  without the addition of a polymer provided  $72\%$  arsenate removal. As is the case with ferric chloride and ferric sulfate, arsenate removal efficiency generally increased with increasing the concentrations of cationic, anionic, and nonionic polyelectrolyte content to  $3 \text{ mg L}^{-1}$  for cationic and nonionic and  $6 \text{ mg L}^{-1}$  for anionic polyelectrolyte. But, when the cationic and nonionic polyelectrolyte dose rose above  $3 \text{ mg L}^{-1}$  arsenate removal efficiency did not change. The percent arsenate removal reached a maximum level at a cationic polyelectrolyte concentration of  $3 \text{ mg L}^{-1}$ . Increasing the coagulant aid dose from  $0.6$  to  $3 \text{ mg L}^{-1}$  resulted in an increase in arsenate removal efficiency from  $75$  to  $76\%$ . The addition of anionic and nonionic polyelectrolyte provided maximum of  $74\%$  arsenate removal.

### DISCUSSION

Arsenate exists in the anionic form of  $\text{H}_2\text{AsO}_4^-$ ,  $\text{HAsO}_4^{2-}$  or  $\text{AsO}_4^{3-}$  above the pH of  $2$  as shown in Figure 5. Therefore, the addition of coagulants facilitates the removal of arsenate from the aqueous stream, by converting the soluble arsenate species

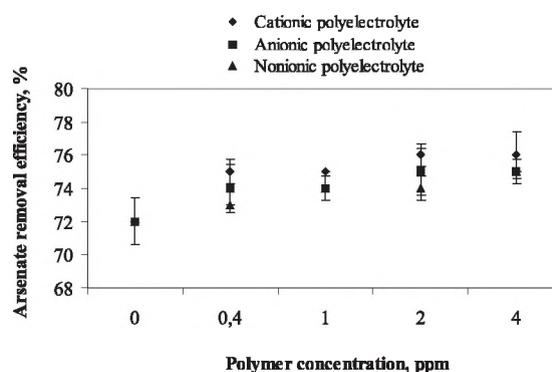


Fig 3. The effect of polyelectrolyte addition on the performance of the coagulation process with  $\text{Fe}_2(\text{SO}_4)_3$  ( $40 \text{ mg L}^{-1}$ )

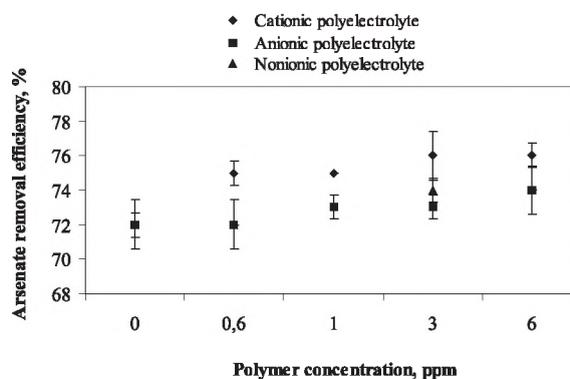


Fig 4. The effect of polyelectrolyte addition on the performance of the coagulation process with  $\text{Fe}_2\text{SO}_4$  ( $60 \text{ mg L}^{-1}$ )

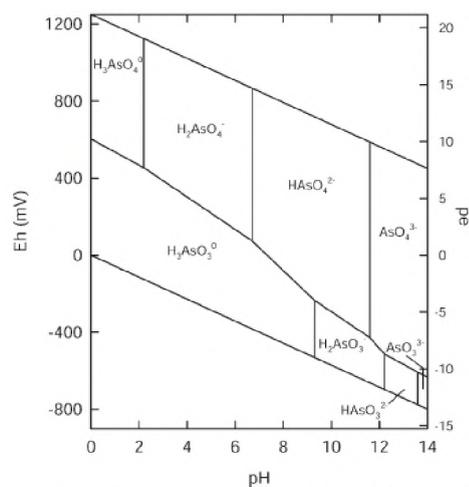
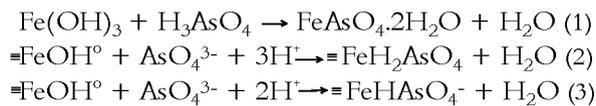


Fig 5. The Eh-pH diagram for inorganic arsenic (Smedley and Kinniburgh 2002).

into insoluble products (McNeill and Edwards 1997, Smedley and Kinniburgh 2002). Our findings showed that the percent arsenate removal increased with the increase of the ferric chloride, ferric sulfate,

and ferrous sulfate dose due to the chemical destabilization of the colloidal particles by neutralizing the charges that keeps them apart. In the coagulation-flocculation process using ferric salts, ferric hydroxide micro-flocs are formed rapidly. The micro-particles and negatively charged ions are attached to the flocs by electrostatic attachment. The possible reactions of arsenate with hydrous iron oxide are shown below where (=FeOH<sup>o</sup>) represents oxide surface site (Mok and Wai 1994, Hering et al. 1996);



Residual arsenate concentrations decreased below 10 mg L<sup>-1</sup> for the ferric chloride and ferrous sulfate dosage of 30 and 80 mg L<sup>-1</sup>, respectively. However, the maximum arsenate removal efficiency of 74% was achieved at an Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> concentration of around 40 mg L<sup>-1</sup>. Therefore ferric chloride proved to be a very efficient coagulant in the removal of arsenate from water at a relatively lower dosage as compared with ferric sulfate and ferrous sulfate. The results obtained from the present study are in agreement with the values given in literature. Pande et al. (1997) investigated arsenic removal by coagulation using ferric chloride. For 500 and 14 µg L<sup>-1</sup> of initial arsenic concentrations, 50 and 30 mg L<sup>-1</sup> of ferric chloride was able to lower the arsenic levels to a permissible limit, respectively. Furthermore in most of the previous studies the amount of coagulants applied were similar in concentrations applied with this study (Edwards 1994, Hering et al. 1997). However, Zouboulis and Katsoyiannis (2002) have used 4-11 mg L<sup>-1</sup> of coagulant concentration and reduced the arsenic concentration below 10 µg L<sup>-1</sup> by applying the combination of pipe flocculation followed by direct filtration. Therefore they have achieved a significant improvement in the traditional coagulation treatment method. Several studies have been reported that arsenic removal from drinking water by coagulation with ferric chloride is more effective than other coagulants such as ferric sulfate, ferrous sulfate, and aluminum sulfate (Han et al. 2002, Wickramasinghe et al. 2004).

This study has also shown that ferric chloride provides minimum and acceptable residual iron

concentration (175 µg L<sup>-1</sup>) after sedimentation and filtration according to Turkish Standards (200 µg L<sup>-1</sup>). Maximum residual iron is measured as about 1400 µg L<sup>-1</sup> for ferrous sulfate. It appears that residual iron increases with increasing ferric chloride and ferrous sulfate concentration and decreasing residual arsenic. Excess doses of ferric chloride and ferrous sulfate resulted in high residual iron and the highest residual iron was found at the FeCl<sub>3</sub> and FeSO<sub>4</sub> concentration of 40 and 90 mg L<sup>-1</sup>, respectively. Residual iron concentration was proportional to arsenate removal in the case of using ferric sulfate as a coagulant. Consequently, residual iron concentration was affected by residual arsenic concentration besides coagulant dose. Experiments by Karcher et al. (1999) were carried out to optimize removal by varying the ferric chloride coagulant dose (3-9 mg L<sup>-1</sup> Fe) and pH value (5.5-8.0) in raw fresh water which contained an average arsenic concentration of 0.44 mg L<sup>-1</sup>. After coagulation-flocculation and sedimentation, the residual concentration of arsenic was 0.035-0.369 mg L<sup>-1</sup> and the residual concentration of Fe was between 14.4% and nearly 100% of the original dose, depending mainly on the dose.

Arsenate removal efficiency was improved by the addition of the organic polymers. Because the addition of cationic, anionic, or nonionic polyelectrolyte enhanced the efficiency of the method, the percentage of arsenate removal was found to be higher than the removal achieved without them. Wickramasinghe et al. (2004) investigated arsenic removal by coagulation and filtration using ferric chloride or ferric sulfate as a coagulant and a cationic polymer as a coagulant aid. Their results indicated that the addition of a cationic polyelectrolyte has little effect on the residual arsenic concentration. Zouboulis and Katsoyiannis (2002) also determined the efficiency of coagulation-pipe flocculation for arsenic removal and investigated the main parameters affecting arsenic removal such as the type and dosage of the coagulant (ferric chloride or alum) and the type and dosage of the polyelectrolyte (cationic or anionic). Their experimental results indicated that the method was efficient for both iron and alum coagulation and both types of polymers (cationic or anionic) were found to increase the overall efficiency of the treatment method.

All types of polymers increased the removal

efficiency of the treatment method in our study. But, the application of a cationic polyelectrolyte was more effective than anionic and nonionic polyelectrolytes. This can be attributed to the fact that these polymers increase the cationic character of the inorganic coagulant, resulting in the formation of solids, which are more likely to adsorb oxyanions (arsenic) (Grohmann 1985, Zouboulis and Katsoyiannis 2002). The application of polyelectrolytes resulted in the formation of greater particles, faster floc formation, and produce denser and stronger flocs (Chen et al. 1994, Wang et al. 2005). Percent arsenate removal reached a

maximum level at cationic polyelectrolyte concentration of 2.5, 2, and 3 mg L<sup>-1</sup> for ferric chloride, ferric sulfate, and ferrous sulfate, respectively.

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